

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

MIYAKI, Yukio, et al.

Continuation of Appln. No.: 08/981,011

Group Art Unit: not yet assigned

Confirmation No.: not yet assigned

Examiner: not yet assigned

Filed: January 17, 2002

For: NONAQUEOUS SECONDARY BATTERY

PRELIMINARY AMENDMENT

Commissioner for Patents  
Washington, D.C. 20231

Sir:

Prior to examination, please amend the above-identified application as follows:

IN THE SPECIFICATION:

**Please amend the specification as follows.**

**Amend the specification by inserting before the first paragraph:**

This is a continuation of Application No. 08/981,011 filed December 24, 1997, which is a Continuation Application of PCT Application No. PCT/JP96/01788 filed June 27, 1996; the above noted prior applications are all hereby incorporated by reference.

**Page 2, please replace the paragraph at line 7 with the following:**

Brief Summary of the Invention

**Page 3, please insert the following paragraphs before the first full paragraph:**

Brief Description of the Drawings:

Fig. 1 illustrates a cross section of a cylindrical battery used in the Examples.

The numerical references used in the Figure indicate the following members:

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CONTINUATION OF U.S. Appln. No. 08/981,011

- 8     ...     Positive sheet electrode
- 9     ...     Negative sheet electrode
- 10    ...     Separator
- 11    ...     Battery case
- 12    ...     Cap
- 13    ...     Gasket
- 14    ...     Explosion-proof valve

Detailed Description of the Invention:

**Page 4, please replace the paragraph bridging pages 4 and 5 with the following:**

The protective layer according to the present invention includes an electrically insulating protective layer, an electrically conducting protective layer, an alkali metal salt- or alkaline earth metal salt-containing protective layer, and an organic fine particles-containing protective layer. These protective layers will be explained below.

The protective layer of the invention comprises at least one layer and may be composed of plural layers of a same or different kinds. The protective layer preferably has a thickness of 1 to 40  $\mu\text{m}$ , more preferably 2 to 30  $\mu\text{m}$ .

[Insulating Protective Layer]

The electrically insulating protective layer used in the invention is a layer having substantially no electron conductivity, i.e., an insulating layer. Where the insulating protective layer comprises a plurality of layers, at least the outermost layer should be insulating. It is not desirable for the protective layer containing particles to melt or form another film at 300°C or less. The protective layer preferably contains insulating

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organic or inorganic fine particles. The fine particles preferably have a particle size of 0.1 to 20  $\mu\text{m}$ , particularly 0.2 to 15  $\mu\text{m}$ .

Preferred organic fine particles are powdered cross-linked latex or fluorine resins. Those having a glass transition point of 250 to 350°C and undergoing no decomposition nor film formation are preferred. Fine particles of Teflon are more preferred.

Examples of inorganic fine particles include carbides, silicides, nitrides, sulfides or oxides or metals or

**Page 6, please replace the second full paragraph and the paragraph bridging pages 6 and 7 with the following:**

The electrically conducting protective layer for use in the invention comprises water-insoluble fine conductive particles and a binder. The binder to be used can be the one used in formation of an electrode material mixture hereinafter described. The content of the fine conductive particles in the conducting protective layer is preferably 2.5 to 96% by weight, more preferably 5 to 95 % by weight, particularly preferably 10 to 93% by weight.

Examples of the water-insoluble fine conductive particles include metals, metal oxides, metallic fibers, carbon fiber, carbon black, and graphite. They preferably have a water solubility of not higher than 100 ppm, and are more preferably insoluble in water. Of these water-insoluble fine conductive particles preferred are those having low reactivity to alkali metals, especially lithium. Metal particles and carbon particles are more preferred. It is preferable for the element constituting the particles to have an electrical resistivity of not higher than  $5 \times 10^9 \Omega \cdot \text{m}$  at 20 °C.

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CONTINUATION OF U.S. Appl. No. 08/981,011**

**Page 7, please replace the first full paragraph with the following:**

Examples of the fine metal particles preferably include those having low reactivity to lithium, i.e, those hardly forming an alloy with lithium, such as copper, nickel, iron, chromium, molybdenum, titanium, tungsten, and tantalum. The fine metal particles can be acicular, columnar, tabular or lumpy, preferably with a maximum diameter of 0.02 to 20  $\mu\text{m}$ , particularly 0.1 to 10  $\mu\text{m}$ . It is desirable for the fine metal particles not to be oxidized to an excessive degree on their surface. If they have an oxidized surface, they are preferably heat-treated in a reducing atmosphere.

**Page 8, please replace the second full paragraph with the following:**

The alkali metal salt- or alkaline earth metal salt-containing protective layer according to the present invention contains fine particles of water-insoluble or sparingly water-soluble alkali metal salt or alkaline earth metal salt (exclusive of chalcogenides) and a binder. These fine particles preferably have a particle size of 0.02 to 20  $\mu\text{m}$ , particularly 0.05 to 10  $\mu\text{m}$ .

**Page 55, please delete the paragraph beginning at line 23 to page 56, line 9.**

**Page 59, please replace the first full paragraph with the following:**

The dried positive sheet electrode (8), a finely porous polypropylene film separator (CELGARD® 2400) (10), the dried negative sheet electrode (9), and the separator (10) were laid one on another in this order and rolled up into a cylinder by means of a winder.

**Page 74, please replace the first line under the heading "Open Circuit Voltage After 3 Weeks" in Table 3 with the following:**

0.99

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CONTINUATION OF U.S. Appl. No. 08/981,011

IN THE CLAIMS:

Please cancel claims 1-32, 34, 38, 54 and 55 without prejudice or disclaimer.

Please enter the following amended claims:

33. (amended) A nonaqueous secondary battery comprising a positive electrode containing a material capable of reversibly intercalating and deintercalating lithium, a negative electrode containing a composite oxide containing tin represented by formula (3):



wherein  $\text{M}^3$  represents at least two elements selected from the group consisting of Al, B, P, and Si;  $\text{M}^4$  represents at least one element selected from the group consisting of elements of groups 1 to 3 of the Periodic Table, and halogen elements;  $c$  represents a number of from 0.2 to 2;  $d$  represents a number of from 0.01 to 1; provided that  $0.2 < c + d < 2$ ; and  $t$  represents a number of from 1 to 6, a nonaqueous electrolyte containing a lithium salt, and a separator, wherein at least one of the negative electrode and positive electrode has at least one protective layer.

39. (amended) The nonaqueous secondary battery as claim 33, wherein said protective layer comprises fine particles of water-insoluble or sparingly water-soluble alkali metal salt or alkaline earth metal salt, and at least two kinds of binders.

40. (amended) The nonaqueous secondary battery as in claim 33, wherein said protective layer has substantially no electrical conductivity.

41. (amended) The nonaqueous secondary battery as in claim 33, wherein said protective layer has electrical conductivity.

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42. (amended) The nonaqueous secondary battery as in claim 33, wherein said protective layer contains particles having substantially no electrical conductivity.

43. (amended) The nonaqueous secondary battery as in claim 33, wherein said protective layer contains particles which are electrically conductive.

44. (amended) The nonaqueous secondary battery as in claim 33, wherein said protective layer contains particles which are inorganic chalcogenide particles.

45. (amended) The nonaqueous secondary battery as in claim 44, wherein said inorganic chalcogenide particles contain at least one inorganic oxide selected from the group consisting of oxide of sodium, potassium, magnesium, calcium, strontium, zirconium, aluminum, and silicon.

47. (amended) The nonaqueous secondary battery as in claim 33, wherein said protective layer contains electrically conductive particles selected from the group consisting of metal powders and carbon particles.

48. (amended) The nonaqueous secondary battery as in claim 47, wherein said electrically conductive particles are carbon particles.

49. (amended) The nonaqueous secondary battery as in claim 39, wherein said water-insoluble or sparingly water-soluble fine particles contained in said protective layer are in an alkali metal salt.

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**PRELIMINARY AMENDMENT**  
**CONTINUATION OF U.S. Appln. No. 08/981,011**

50. (amended) The nonaqueous secondary battery as in claim 39, wherein said water-insoluble or sparingly water-soluble fine particles contained in said protective layer are in an alkali earth metal salt.

51. (amended) The nonaqueous secondary battery as in claim 49, wherein said fine particles are a lithium salt.

52. (amended) The nonaqueous secondary battery as in claim 33, wherein said protective layer has a thickness of from 1  $\mu\text{m}$  to 40  $\mu\text{m}$ .

53. (amended) The nonaqueous secondary battery as in claim 33, wherein said protective layer contains electrically conductive particles in an amount of from 2.5 to 96% by weight.

56. (amended) The nonaqueous secondary battery as in claim 33, wherein said nonaqueous electrolyte contains at least one carbonic ester.

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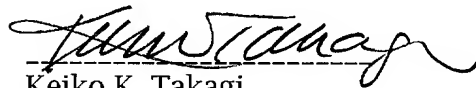
PRELIMINARY AMENDMENT  
CONTINUATION OF U.S. Appln. No. 08/981,011

REMARKS

Claims 33, 35-37, 39-53 and 56 are all the claims pending in this application. The specification has been amended to address informalities raised by the Examiner in the parent application. In addition, support for the amendments characterizing the particles as "fine" can be found, for example, at page 7, lines 9-12 and page 8, line 1 and lines 15-17 of the present specification. With respect to the amendment to Table 3, Applicants submit that "3.99" was inadvertently typed instead of "0.99". In this regard, Applicants submit herewith a Rule 132 Declaration showing that the error is necessarily and inevitably inherent in the original disclosure. The Rule 132 Declaration states the steps carried out to prepare Battery AS, and the evaluation of Battery AS, as set forth in the specification at pages 71-73. Accordingly, the Declaration shows that "3.99" is a typographical error.

Entry and consideration of this Amendment is respectfully requested.

Respectfully submitted,



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Registration No. 47,121

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Date: January 17, 2002



APPENDIX

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION:

The specification is changed as follows:

Page 1, before the first paragraph, the following has been inserted.

This is a continuation of Application No. 08/981,011 filed December 24, 1997, which is a Continuation Application of PCT Application No. PCT/JP96/01788 filed June 27, 1996; the above noted prior applications are all hereby incorporated by reference.

Page 2, the paragraph at line 7:

[Disclosure] Brief Summary of the Invention

Page 3, the following paragraphs were inserted before the first full paragraph:

Brief Description of the Drawings:

Fig. 1 illustrates a cross section of a cylindrical battery used in the Examples.

The numerical references used in the Figure indicate the following members:

- 8        ...        Positive sheet electrode
- 9        ...        Negative sheet electrode
- 10      ...        Separator
- 11      ...        Battery case
- 12      ...        Cap
- 13      ...        Gasket
- 14      ...        Explosion-proof valve

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Detailed Description of the Invention:

**Page 4, the paragraph bridging pages 4 and 5:**

The protective layer according to the present invention includes an electrically insulating protective layer, an electrically conducting protective layer, an alkali metal salt- or alkaline earth metal salt-containing protective layer, and an organic fine particles-containing protective layer. These protective layers will be explained below.

The protective layer of the invention comprises at least one layer and may be composed of plural layers of a same or different kinds. The protective layer preferably has a thickness of 1 to 40  $\mu\text{m}$ , more preferably 2 to 30  $\mu\text{m}$ .

[Insulating Protective Layer]

The electrically insulating protective layer used in the invention is a layer having substantially no electron conductivity, i.e., an insulating layer. Where the insulating protective layer comprises a plurality of layers, at least the outermost layer should be insulating. It is not desirable for the protective layer containing particles to melt or form another film at 300°C or less. The protective layer preferably contains insulating organic or inorganic fine particles. The fine particles preferably have a particle size of 0.1 to 20  $\mu\text{m}$ , particularly 0.2 to 15  $\mu\text{m}$ .

Preferred organic fine particles are powdered cross-linked latex or fluorine resins. Those having a glass transition point of 250 to 350°C and undergoing no decomposition nor film formation are preferred. Fine particles of Teflon are more preferred.

Examples of inorganic fine particles include carbides, silicides, nitrides, sulfides or oxides or metals or

**Page 6, the second full paragraph and the paragraph bridging pages 6 and 7:**

The electrically conducting protective layer for use in the invention comprises water-insoluble fine conductive particles and a binder. The binder to be used can be the one used in formation of an electrode material mixture hereinafter described. The content of the fine conductive particles in the conducting protective layer is preferably 2.5 to 96% by weight, more preferably 5 to 95 % by weight, particularly preferably 10 to 93% by weight.

Examples of the water-insoluble fine conductive particles include metals, metal oxides, metallic fibers, carbon fiber, carbon black, and graphite. They preferably have a water solubility of not higher than 100 ppm, and are more preferably insoluble in water. Of these water-insoluble fine conductive particles preferred are those having low reactivity to alkali metals, especially lithium. Metal particles and carbon particles are more preferred. It is preferable for the element constituting the particles to have an electrical resistivity of not higher than  $5 \times 10^9$  [Q·m]  $\Omega \cdot m$  at 20 °C.

**Page 7, the first full paragraph:**

Examples of the fine metal particles preferably [includes] include those having low reactivity to lithium, i.e, those hardly forming an alloy with lithium, such as copper, nickel, iron, chromium, molybdenum, titanium, tungsten, and tantalum. The fine metal particles can be acicular, columnar, tabular or lumpy, preferably with a maximum diameter of 0.02 to 20  $\mu m$ , particularly 0.1 to 10  $\mu m$ . It is desirable for the fine metal particles not to be oxidized to an excessive degree on their surface. If they have an oxidized surface, they are preferably heat-treated in [an] a reducing atmosphere.

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**Page 8, the second full paragraph:**

The alkali metal salt- or alkaline earth metal salt-containing protective layer according to the present invention contains fine particles of water-insoluble or sparingly water-soluble alkali metal salt or alkaline earth metal salt (exclusive of chalcogenides) and a binder. These fine particles preferably have a particle size of 0.02 to 20  $\mu\text{m}$ , particularly 0.05 to 10  $\mu\text{m}$ .

**Page 55, the paragraph beginning at line 23 to page 56, line 9 has been deleted.**

**Page 59, the first full paragraph:**

The dried positive sheet electrode (8), a finely porous polypropylene film separator ([Cell Guard] CELGARD® 2400) (10), the dried negative sheet electrode (9), and the separator (10) were laid one on another in this order and rolled up into a cylinder by means of a winder.

**Page 74, the first line under the heading "Open Circuit Voltage After 3 Weeks" in Table 3:**

[3.99] 0.99

**IN THE CLAIMS:**

**Claims 1-32, 34, 38, 54 and 55 have been canceled.**

**The claims have been amended as follows:**

33. (amended) A nonaqueous secondary battery comprising a positive electrode containing a material capable of reversibly intercalating and deintercalating lithium, a negative electrode containing [at least one oxide selected from the group consisting of a metal or semimetal oxide belonging to the groups 13 to 15 of the

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Periodic Table which is capable of reversibly intercalating and deintercalating lithium]  
a composite oxide containing tin represented by formula (3):



wherein M<sup>3</sup> represents at least two elements selected from the group consisting of Al, B, P, and Si; M<sup>4</sup> represents at least one element selected from the group consisting of elements of groups 1 to 3 of the Periodic Table, and halogen elements; c represents a number of from 0.2 to 2; d represents a number of from 0.01 to 1; provided that 0.2 < c + d < 2; and t represents a number of from 1 to 6, a nonaqueous electrolyte containing a lithium salt, and a separator, wherein at least one of the negative electrode and positive electrode has at least one protective layer.

39. (amended) The nonaqueous secondary battery as [in any one of claims 35 to 37] claim 33, wherein said protective layer comprises fine particles of water-insoluble or sparingly water-soluble[,] alkali metal salt or alkaline earth metal salt, and [a binder] at least two kinds of binders.

40. (amended) The nonaqueous secondary battery as in [any one of claims 35 to 39] claim 33, wherein said protective layer has substantially no electrical conductivity.

41. (amended) The nonaqueous secondary battery as in [any one of claims 35 to 39] claim 33, wherein said protective layer has electrical conductivity.

PRELIMINARY AMENDMENT  
CONTINUATION OF U.S. Appln. No. 08/981,011

42. (amended) The nonaqueous secondary battery as in claim [41] 33, wherein said protective layer contains particles having substantially no electrical conductivity.

43. (amended) The nonaqueous secondary battery as in claim [41 or 42] 33, wherein said protective layer contains particles which are [an] electrically conductive [particle].

44. (amended) The nonaqueous secondary battery as in claim [38 or 39] 33, wherein said protective layer contains particles which are [an] inorganic chalcogenide [particle] particles.

45. (amended) The nonaqueous secondary battery as in claim 44, wherein said inorganic chalcogenide [particle] particles [contains] contain at least one inorganic oxide selected from the group consisting of oxide of sodium, potassium, magnesium, calcium, strontium, zirconium, aluminum, and silicon.

47. (amended) The nonaqueous secondary battery as in claim [38 or 39] 33, wherein said protective layer contains electrically conductive [particle] particles [is at least one particle] selected from the group consisting of metal powders and carbon particles.

48. (amended) The nonaqueous secondary battery as in claim 47, wherein said electrically conductive [particle is a] particles are carbon [particle] particles.

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CONTINUATION OF U.S. Appln. No. 08/981,011

49. (amended) The nonaqueous secondary battery as in claim [38 or] 39, wherein said water-insoluble or sparingly water-soluble fine particles contained in said protective layer are in an alkali metal salt.

50. (amended) The nonaqueous secondary battery as in claim [38 or] 39, wherein said water-insoluble or sparingly water-soluble fine particles contained in said protective layer are in an alkali earth metal salt.

51. (amended) The nonaqueous secondary battery as in claim [50] 49, wherein said fine particles are a lithium salt.

52. (amended) The nonaqueous secondary battery as in [any one of claims] claim 33 [to 51], wherein said protective layer has a thickness of from 1  $\mu\text{m}$  to 40  $\mu\text{m}$ .

53. (amended) The nonaqueous secondary battery as in claim [52] 33, wherein said protective layer contains electrically conductive particles in an amount of from 2.5 to 96% by weight.

56. (amended) The nonaqueous secondary battery as in [any one of claims] claim 33 [to 55], wherein said nonaqueous electrolyte contains at least one carbonic ester.

10045706, 01.4.2010

AMENDMENT

(Amendment under the PCT Article 34)

Director-General, Patent Office:

1. Indication of International Application

PCT/JP96/01788

2. Applicant

Name: Fuji Photo Film Co., Ltd.  
Address: 210, Nakanuma, Minami-Ashigara-Shi  
KANAGAWA 250-01 Japan  
Nationality: Japan  
Address: Japan

3. Agent

Name: 7387 Patent Attorney HAGINO Taira  
Address: Eikoh Patent Office,  
28th Floor, ARK MORI Building, 12-32,  
Akasaka 1-Chome, Minato-ku, TOKYO 107  
JAPAN

4. Date of Amendment Order

Voluntary

5. Object of Amendment Order

Specification

Claims

6. Contents of Amendment

The amendment is as follows



(1) Japanese text, at page 1, lines 22-24

(English specification, at page 2, lines 10-15)

Pleas amend

"(1) A nonaqueous secondary battery comprising a positive electrode and a negative electrode both containing a material capable of reversibly intercalating and deintercalating lithium, and a nonaqueous electrolyte containing a lithium salt, wherein at least one of the negative electrode and positive electrode has at least one protective layer." to

-- (1) A nonaqueous secondary battery comprising a positive electrode and a negative electrode both containing a material capable of reversibly intercalating and deintercalating lithium, and a nonaqueous electrolyte containing a lithium salt, wherein at least one of the negative electrode and positive electrode has at least one protective layer containing at least one inorganic oxide selected from the group consisting of alumina, silicon dioxide and zirconia. --.

(2) Japanese text, at page 1, lines 25-27.

(English specification, at page 2, lines 16-22)

Please amend

"(2) A nonaqueous secondary battery comprising a positive electrode and a negative electrode both containing a material capable of reversibly intercalating and

deintercalating lithium, and a nonaqueous electrolyte containing a lithium salt, and a separator, wherein at least one of the negative electrode and positive electrode has at least one protective layer." to

-- (2) A nonaqueous secondary battery comprising a positive electrode and a negative electrode both containing a material capable of reversibly intercalating and deintercalating lithium, and a nonaqueous electrolyte containing a lithium salt, wherein at least one of the negative electrode and positive electrode has at least one protective layer containing organic fine particles. --.

(3) Japanese text, at page 2, lines 1-4.

(English specification, at page 2, line 3 from the bottom to page 3, line 4)

Please amend

"(3) A nonaqueous secondary battery comprising a positive electrode containing a material capable of reversibly intercalating and deintercalating lithium, a negative electrode mainly comprising a metal or semimetal oxide, a nonaqueous electrolyte containing a lithium salt, and a separator, wherein at least one of the negative electrode and positive electrode has at least one protective layer." to

-- (3) A nonaqueous secondary battery comprising a positive electrode containing a material capable of

reversibly intercalating and deintercalating lithium, a negative electrode containing at least one oxide selected from the group consisting of a metal or semimetal oxide belonging to the groups 13 to 15 of the Periodic Table which is capable of reversibly intercalating and deintercalating lithium, a nonaqueous electrolyte containing a lithium salt, and a separator, wherein at least one of the negative electrode and positive electrode has at least one protective layer. --.

(4) Japanese text, claim 1, lines 2-3.

(English text, claim 1, lines 6-7)

Please amend "at least one protective layer" to -- at least one protective layer containing at least one inorganic oxide selected from the group consisting of alumina, silicon dioxide and zirconia --.

(5) Japanese text, claim 2.

(English specification, claim 2)

Please delete claim 2.

(6) Japanese text, claim 3

(English specification, claim 3)

Please amend

"3. A nonaqueous secondary battery comprising a positive electrode containing a material capable of

reversibly intercalating and deintercalating lithium, a negative electrode mainly comprising a metal or semimetal oxide, a nonaqueous electrolyte containing a lithium salt, and a separator, wherein at least one of said negative electrode and positive electrode has at least one protective layer." to

-- 3. A nonaqueous secondary battery comprising a positive electrode and a negative electrode both containing a material capable of reversibly intercalating and deintercalating lithium and a nonaqueous electrolyte containing a lithium salt, wherein at least one of said negative electrode and positive electrode has at least one protective layer containing organic fine particles. --.

(7) Japanese text, claim 4.

(English specification, claim 4)

Please delete claim 4.

(8) Japanese text, claim 5

(English specification, claim 5)

Please delete claim 5.

(9) Japanese text, claim 6

(English specification, claim 6)

Please delete claim 6.

(10) Japanese text, claim 7

(English specification, claim 7)

Please delete claim 7.

(11) Japanese text, claim 8

(English specification, claim 8)

Please delete claim 8.

(12) Japanese text, claim 9

(English specification, claim 9)

Please delete claim 9.

(13) Japanese text, claim 10, line 2

(English specification, claim 10, lines 1-2)

Please amend "any one of claims 4 to 6" to -- claim  
3 --.

(14) Japanese text, claim 11, lines 1-2

(English specification, claim 11, lines 1-2)

Please amend "any one of claims 4 to 10" to -- claim  
3 or 10 --.

(15) Japanese text, claim 12, lines 1-2

(English specification, claim 12, lines 1-2)

Please amend "any one of claims 4 to 10" to -- claim  
3, 10 or 11 --.

(16) Japanese text, claim 15, line 2

(English specification, claim 15, lines 1-2)

Please amend "any one of claims 7 to 10" to -- claim  
10 --.

(17) Japanese text, claim 18

(English specification, claim 18)

Please delete claim 18.

(18) Japanese text, claim 19

(English specification, claim 19)

Please delete claim 19.

(19) Japanese text, claim 20

(English specification, claim 20)

Please delete claim 20.

(20) Japanese text, claim 21

(English specification, claim 21)

Please delete claim 21.

(21) Japanese text, claim 22

(English specification, claim 22)

Please delete claim 22.

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(22) Japanese text, claim 23, line 1

(English specification, claim 23, lines 1-2)

Please amend "claim 9 or 10" to -- claim 10 --.

(23) Japanese text, claim 24, line 1

(English specification, claim 24, lines 1-2)

Please amend "claim 9 or 10" to -- claim 10 --.

(24) Japanese text, claim 25, line 1

(English specification, claim 25, lines 1-2)

Please amend "claim 9 or 10" to -- claim 10 --.

(25) Japanese text, claim 26, line 2

(English specification, claim 26, line 2)

Please amend "claims 1 to 25" to -- claims 3, 10 to 17 and 23 to 25 --.

(26) Japanese text, claim 28, line 3

(English specification, claim 28, line 2)

Please amend "claims 1 to 27" to -- claims 3, 10 to 17 and 23 to 27 --.

(27) Japanese text, claim 32, line 2

(English specification, claims 32, line 2)

Please amend "claims 1 to 31" to -- claims 3, 10 to 17 and 23 to 31 --.

(28) Please add the following claim 33.

33. . A nonaqueous secondary battery comprising a positive electrode containing a material capable of reversibly intercalating and deintercalating lithium, a negative electrode containing at least one oxide selected from the group consisting of a metal or semimetal oxide belonging to the groups 13 to 15 of the Periodic Table which is capable of reversibly intercalating and deintercalating lithium, a nonaqueous electrolyte containing a lithium salt, and a separator, wherein at least one of the negative electrode and positive electrode has at least one protective layer.

(29) Please add the following claim 34.

34. The nonaqueous secondary battery as in claim 33, wherein said negative electrode is a composite oxide containing tin.

(30) Please add the following claim 35.

35. The nonaqueous secondary battery as in claim 33, wherein said protective layer is formed on both of the positive electrode and negative electrode.

(31) Please add the following claim 36.

36. The nonaqueous secondary battery as in claim 33, wherein said protective layer is formed on the negative



electrode.

(32) Please add the following claim 37.

37. The nonaqueous secondary battery as in claim 33, wherein said protective layer is formed on the positive electrode.

(33) Please add the following claim 38.

38. The nonaqueous secondary battery as in any one of claims 35 to 37, wherein said protective layer comprises water-insoluble particles and a binder.

(34) Please add the following claim 39.

39. The nonaqueous secondary battery as in any one of claims 35 to 37, wherein said protective layer comprises particles of water-insoluble or sparingly water-soluble, alkali metal salt or alkaline earth metal salt, and a binder.

(35) Please add the following claim 40.

40. The nonaqueous secondary battery as in any one of claims 35 to 39, wherein said protective layer has substantially no electrical conductivity.

(36) Please add the following claim 41.

41. The nonaqueous secondary battery as in any one of claims 35 to 39, wherein said protective layer has

electrical conductivity.

(37) Please add the following claim 42.

42. The nonaqueous secondary battery as in claim 41, wherein said protective layer contains particles having substantially no electrical conductivity.

(38) Please add the following claim 43.

43. The nonaqueous secondary battery as in claim 41 or 42, wherein said protective layer contains particles which are an electrically conductive particle.

(39) Please add the following claim 44.

44. The nonaqueous secondary battery as in claim 38 or 39, wherein said protective layer contains particles which are an inorganic chalcogenide particle.

(40) Please add the following claim 45.

45. The nonaqueous secondary battery as in claim 44, wherein said inorganic chalcogenide particle contains at least one oxide selected from the group consisting of oxide of sodium, potassium, magnesium, calcium, strontium, zirconium, aluminum, and silicon.

(41) Please add the following claim 46.

46. The nonaqueous secondary battery as in claim 45,

wherein said inorganic oxide is selected from the group consisting of alumina, silicon dioxide and zirconia.

(42) Please add the following claim 47.

47. The nonaqueous secondary battery as in claim 38 or 39, wherein said electrically conductive particle is at least one particle selected from the group consisting of metal powders and carbon particles.

(43) Please add the following claim 48.

48. The nonaqueous secondary battery as in claim 47, wherein said electrically conductive particle is a carbon particle.

(44) Please add the following claim 49.

49. The nonaqueous secondary battery as in claim 38 or 39, wherein said water-insoluble or sparingly water-soluble particles contained in said protective layer are an alkali metal salt.

(45) Please add the following claim 50.

50. The nonaqueous secondary battery as in claim 38 or 39, wherein said water-insoluble or sparingly water-soluble particles contained in said protective layer are an alkaline earth metal salt.

(46) Please add the following claim 51.

51. The nonaqueous secondary battery as in claim 50, wherein said particles are a lithium salt.

(47) Please add the following claim 52.

52. The nonaqueous secondary battery as in any one of claims 33 to 51, wherein said protective layer has a thickness of from 1  $\mu\text{m}$  to 40  $\mu\text{m}$ .

(48) Please add the following claim 53.

53. The nonaqueous secondary battery as in claim 52, wherein said protective layer contains electrically conductive particles in an amount of from 2.5 to 96% by weight.

(49) Please add the following claim 54.

54. The nonaqueous secondary battery as in claim 53, wherein said composite oxide containing tin is represented by formula (1):



wherein  $\text{M}^1$  represents two or more elements selected from the group consisting of Al, B, P, Si, Ge, elements of groups 1 to 3 of the Periodic Table, and halogen elements; a represents a number of from 0.2 to 2; and t represents a number of from 1 to 6.

(50) Please add the following claim 55.

55. The nonaqueous secondary battery as in claim 30, wherein said composite oxide containing tin is represented by formula (3):



wherein  $\text{M}^3$  represents at least two elements selected from the group consisting of Al, B, P, and Si;  $\text{M}^4$  represents at least one element selected from the group consisting of elements of groups 1 to 3 of the Periodic Table, and halogen elements; c represents a number of from 0.2 to 2; d represents a number of from 0.01 to 1; provided that  $0.2 < c + d < 2$ ; and t represents a number of from 1 to 6.

(51) Please add the following claim 56.

56. The nonaqueous secondary battery as in any one of claims 33 to 55, wherein said nonaqueous electrolyte contains at least one carbonic ester.

#### 7. List of Attached Papers

Japanese text, pages 1 and 2, and all of claims  
(English specification, pages 1 to 3, and all of  
claims)

CLAIMS:

1. (Amended) A nonaqueous secondary battery comprising a positive electrode and a negative electrode both containing a material capable of reversibly intercalating and deintercalating lithium, and a nonaqueous electrolyte containing a lithium salt, wherein at least one of said negative electrode and positive electrode has at least one protective layer containing at least one inorganic oxide selected from the group consisting of alumina, silicon dioxide and zirconia.

2. (deleted)

3. (amended) A nonaqueous secondary battery comprising a positive electrode and a negative electrode both containing a material capable of reversibly intercalating and deintercalating lithium, and a nonaqueous electrolyte containing a lithium salt, wherein at least one of said negative electrode and positive electrode has at least one protective layer containing organic fine particles.

4. (deleted)

5. (deleted)

6. (deleted)

7. (deleted)

8. (deleted)

9. (deleted)

10. (amended) The nonaqueous secondary battery as in claim 3, wherein said protective layer contains organic

fine particles and inorganic fine particles.

11. (amended) The nonaqueous secondary battery as in claim 3 or 10, wherein said protective layer has substantially no electrical conductivity.

12. (amended) The nonaqueous secondary battery as in claim 3, 10 or 11, wherein said protective layer is electrically conductive.

13. The nonaqueous secondary battery as in claim 11, wherein said protective layer contains particles having substantially no electrical conductivity.

14. The nonaqueous secondary battery as in claim 11 or 12, wherein said particles in said protective layer are electrically conductive particles.

15. (amended) The nonaqueous secondary battery as in claim 10, wherein said particles in said protective layer are inorganic chalcogenide particles.

16. The nonaqueous secondary battery as in claim 15, wherein said inorganic chalcogenide particles contain at least one oxide selected from the group consisting of oxides of sodium, potassium, magnesium, calcium, strontium, zirconium, aluminum, and silicon.

17. The nonaqueous secondary battery as in claim 16, wherein the inorganic oxide is alumina, silicon dioxide or zirconia.

18. (deleted)

19. (deleted)

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20. (deleted)

21. (deleted)

22. (deleted)

23. (amended) The nonaqueous secondary battery as in claim 10, wherein said organic fine particles have a minimum film-forming temperature (MFT) of from 80 to 200°C.

24. (amended) The nonaqueous secondary battery as in claim 10, wherein said organic fine particles in said protective layer are polyethylene fine particles.

25. (amended) The nonaqueous secondary battery as in claim 10, wherein said inorganic fine particles in said protective layer are selected from the group consisting of lithium fluoride, silicon carbide, and boron nitride.

26. (amended) The nonaqueous secondary battery as in any one of claims 3, 10 to 17 and 23 to 25, wherein said protective layer has a thickness of from 1 to 40  $\mu\text{m}$ .

27. The nonaqueous secondary battery as in claim 26, wherein said protective layer contains electrically conductive particles in a proportion of 2.5 to 96% by weight.

28. (amended) The nonaqueous secondary battery as in any one of claims 3, 10 to 17 and 23 to 27, wherein the negative electrode material capable of reversibly intercalating and deintercalating lithium contains at least one of oxides of metals and semimetals belonging to the groups 13 to 15 of the Periodic Table.

29. The nonaqueous secondary battery as in claim



28, wherein said negative electrode is a composite oxide containing tin.

30. The nonaqueous secondary battery as in claim 29, wherein said composite oxide containing tin is represented by formula (1):



wherein  $M^1$  represents two or more elements selected from the group consisting of Al, B, P, Si, Ge, elements of groups 1 to 3 of the Periodic Table, and halogen elements;  $a$  represents a number of from 0.2 to 2; and  $t$  represents a number of from 1 to 6.

31. The nonaqueous secondary battery as in claim 30, wherein said composite oxide containing tin is represented by formula (3):



wherein  $M^3$  represents at least two elements selected from the group consisting of Al, B, P, and Si;  $M^4$  represents at least one element selected from the group consisting of elements of groups 1 to 3 of the Periodic Table and halogen elements;  $c$  represents a number of from 0.2 to 2;  $d$  represents a number of from 0.01 to 1; provided that  $0.2 < c + d < 2$ ; and  $t$  represents a number of from 1 to 6.

32. (amended) The nonaqueous secondary battery as in any one of claims 3, 10 to 17 and 23 to 31, wherein said nonaqueous electrolyte contains at least one carbonic ester.

33. (added) A nonaqueous secondary battery

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comprising a positive electrode containing a material capable of reversibly intercalating and deintercalating lithium, a negative electrode containing at least one oxide selected from the group consisting of a metal or semimetal oxide belonging to the groups 13 to 15 of the Periodic Table which is capable of reversibly intercalating and deintercalating lithium, a nonaqueous electrolyte containing a lithium salt, and a separator, wherein at least one of the negative electrode and positive electrode has at least one protective layer.

34. (added) The nonaqueous secondary battery as in claim 33, wherein said negative electrode is a composite oxide containing tin.

35. (added) The nonaqueous secondary battery as in claim 33, wherein said protective layer is formed on both of the positive electrode and negative electrode.

36. (added) The nonaqueous secondary battery as in claim 33, wherein said protective layer is formed on the negative electrode.

37. (added) The nonaqueous secondary battery as in claim 33, wherein said protective layer is formed on the positive electrode.

38. (added) The nonaqueous secondary battery as in any one of claims 35 to 37, wherein said protective layer comprises water-insoluble particles and a binder.

39. (added) The nonaqueous secondary battery as

in any one of claims 35 to 37, wherein said protective layer comprises particles of water-insoluble or sparingly water-soluble, alkali metal salt or alkaline earth metal salt, and a binder.

40. (added) The nonaqueous secondary battery as in any one of claims 35 to 39, wherein said protective layer has substantially no electrical conductivity.

41. (added) The nonaqueous secondary battery as in any one of claims 35 to 39, wherein said protective layer has electrical conductivity.

42. (added) The nonaqueous secondary battery as in claim 41, wherein said protective layer contains particles having substantially no electrical conductivity.

43. (added) The nonaqueous secondary battery as in claim 41 or 42, wherein said protective layer contains particles which are an electrically conductive particle.

44. (added) The nonaqueous secondary battery as in claim 38 or 39, wherein said protective layer contains particles which are an inorganic chalcogenide particle.

45. (added) The nonaqueous secondary battery as in claim 44, wherein said inorganic chalcogenide particle contains at least one oxide selected from the group consisting of oxides of sodium, potassium, magnesium, calcium, strontium, zirconium, aluminum, and silicon.

46. (added) The nonaqueous secondary battery as in claim 45, wherein said inorganic oxide is selected from

the group consisting of alumina, silicon dioxide and zirconia.

47. (added) The nonaqueous secondary battery as in claim 38 or 39, wherein said electrically conductive particle is at lease one particle selected from the group consisting of metal powders and carbon particles.

48. (added) The nonaqueous secondary battery as in claim 47, wherein said electrically conductive particle is a carbon particle.

49. (added) The nonaqueous secondary battery as in claim 38 or 39, wherein said water-insoluble or sparingly water-soluble <sup>fine</sup> particles contained in said protective layer are an alkali metal salt.

50. (added) The nonaqueous secondary battery as in claim 38 or 39, wherein said water-insoluble or sparingly water-soluble <sup>fine</sup> particles contained in said protective layer are an alkaline earth metal salt.

51. (added) The nonaqueous secondary battery as in claim 50, wherein said <sup>fine</sup> particles are a lithium salt.

52. (added) The nonaqueous secondary battery as in any one of claims 33 to 51, wherein said protective layer has a thickness of from 1  $\mu$ m to 40  $\mu$ m.

53. (added) The nonaqueous secondary battery as in claim 52, wherein said protective layer contains electrically conductive particles in an amount of from 2.5 to 96% by weight.

54. (added) The nonaqueous secondary battery as in claim 53, wherein said composite oxide containing tin is represented by formula (1):



wherein  $M^1$  represents two or more elements selected from the group consisting of Al, B, P, Si, Ge, elements of groups 1 to 3 of the Periodic Table, and halogen elements; a represents a number of from 0.2 to 2; and t represents a number of from 1 to 6.

55. (added) The nonaqueous secondary battery as in claim 30, wherein said composite oxide containing tin is represented by formula (3):



wherein  $M^3$  represents at least two elements selected from the group consisting of Al, B, P, and Si;  $M^4$  represents at least one element selected from the group consisting of elements of groups 1 to 3 of the Periodic Table, and halogen elements; c represents a number of from 0.2 to 2; d represents a number of from 0.01 to 1; provided that  $0.2 < c + d < 2$ ; and t represents a number of from 1 to 6.

56. (added) The nonaqueous secondary battery as in any one of claims 33 to 55, wherein said nonaqueous electrolyte contains at least one carbonic ester.